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Thermal Instability of Porous Gold Nanowires

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Due to their high surface-to-volume ratio, porous metal and semiconductor nanowires (NWs) are attracting strong interest as promising devices e.g. in electronics, catalysis and sensorics. Under working conditions, NW devices should stand elevated temperatures, it is thus necessary to investigate their thermal stability.

Theoretical modelling by Nichols and Mullins predicted that, while annealing, solid cylinders with initial radius R_0 break into a row of spheres with diameter $3.78R_0$ spaced by $2\sqrt{2}\pi R_0$ in a process governed by surface diffusion [1]. Recent publications showed that Cu and Au NWs are thermally instable at temperatures much lower than the melting point of their bulk counterpart, and transform into a row of spheres [2,3]. Here we study the thermal instability of porous Au NWs by annealing and scanning electron microscopy (SEM) investigations.

AuAg alloy solid NWs were electrochemically grown in the nanochannels of etched ion-track polycarbonate templates. The membranes were fabricated by irradiation with Au ions (~ GeV) at the UNILAC accelerator and subsequent chemical etching.

Nanowires with length ~ 2 µm and diameter ~ 110 nm were synthesized inside the etched channels using a cyanidic electrolyte containing $KAu(CN)_2$ and $KAg(CN)_2$ in 1:1 ratio, applying a potential U = -1.1V vs. Ag/AgCl reference electrode. Details of the synthesis process are given in [4]. After deposition, the polymer membrane is removed in a dichloromethane solution, and the nanowires were drop-casted on Si wafers.

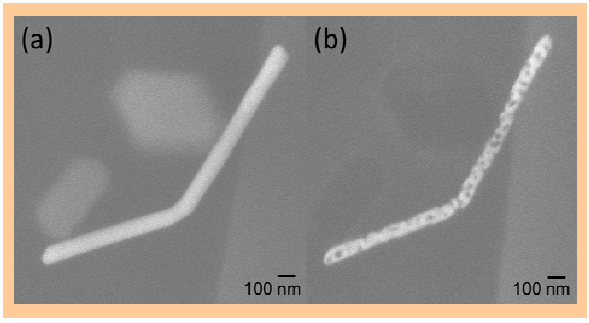

Figure 1: SEM images of a given AuAg NW (a) before and (b) after HNO_3 treatment.

To create porous NWs, the Si wafer with AuAg alloy NWs is dipped for 3h in concentrated nitric acid. Figure 1 depicts the same AuAg alloy NW (a) before and (b) after the nitric acid treatment. In this case, the diameter of the wire is reduced from 105 to 85 nm. Nitric acid dissolves Ag atoms but does not react with Au. The formation of nanoscale porosity can be explained by diffusion of Au adatoms on the NW surface and formation of Au-rich clusters during Ag dissolution [4]. We observe that the porosity and the diameter reduction depend sensitively on the initial Au/Ag composition and NW diameter.

Figure 2 depicts the porous NWs after annealing at 200, 300, and 500 °C for 1h on 290-µm thick silicon wafers (heating rate of 9 °C/min).

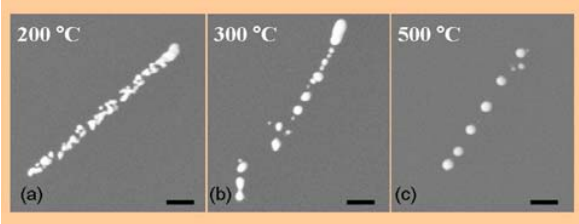

Figure 2: SEM pictures of porous nanowires after 1h annealing at temperatures (a) 200 °C, (b) 300 °C, and (c) 500 °C (Scale bar: 100nm).

Karim et al. previously reported that several µm-long solid Au NWs with diameter ~ 80 nm maintain their cylindrical morphology during 1 hour annealing at temperatures between 200 and 500 °C, and display morphological instabilities after annealing at 600 °C.

In contrast, the morphology of the porous NWs as synthesized in this work starts to change already at 200 °C (Fig. 2a). At higher temperatures, they transform into chains of spheres (Fig. 2b). At 500 °C, spheres with sizes varying between ~ 30 and 120 nm, and inter-sphere separations ranging between 30 and 200 nm are observed (Fig. 2c). Further experiments are currently underway to understand the relation between initial wire porosity and dimensions, and the final morphology of the chain of spheres.

These first results indicate that porous nanostructures undergo morphological transformation at lower temperatures as their solid counterparts due to their higher surface-to-volume ratio. At the same time, this behaviour enables the fabrication of chains of closely spaced nanoparticles interesting for, e.g., surface plasmon investigations.

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